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Instrument Calibration And Measurement Plan For The Poorly Measured/Unmeasured Category of Highly Enriched Uranium at Lawrence Livermore National Laboratory

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Abstract

In partial response to a Department of Energy (DOE) request to evaluate the state of measurements of special nuclear material, Lawrence Livermore National Laboratory (LLNL) evaluated and classified all highly enriched uranium (HEU) metal and oxide items in its inventory. Because of a lack of traceable HEU standards, no items were deemed to fit the category of well measured. A subsequent DOE-HQ sponsored survey by New Brunswick Laboratory resulted in their preparation of a set of certified reference material (CRM) standards for HEU oxide (U_3O_8) that are projected for delivery during September of 1999. However, CRM standards for HEU metal are neither in preparation nor are they expected to be prepared within the foreseeable future. Consequently, HEU metal working standards must be developed if the poorly measured/unmeasured portion of the LLNL inventory is to be reclassified. This paper describes the approach that LLNL will take to (1) develop a set of HEU metal working standards; (2) develop HEU metal and oxide calibration curves for the passive-active neutron (PAN) shuffler that are functions of mass, enrichment, size, and shape; and (3) reclassify the poorly measured/unmeasured inventory through direct measurement or reprocessing of previously archived data.

Background

Measurement Assessment Project

The Measurement Assessment Project (MAP) was initiated in June 1996 at the request of the Department of Energy (DOE) to evaluate the state of measurements of special nuclear material (SNM) across the DOE complex. Each site within the DOE complex was required to state how much material could be classified into each of four categories: well measured, poorly measured/unmeasured, difficult to measure, or non-amenable to measurement.

In partial response to the MAP request, Lawrence Livermore National Laboratory (LLNL) evaluated and classified all highly enriched uranium (HEU) metal and oxide items in inventory. Because of a lack of traceable HEU standards, no items were deemed to fit the category of well measured,

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including the majority of HEU metal items whose book values came from shippers values which were established using destructive analysis data and part weights. Consequently, the classification of LLNL HEU was as follows:

_ poorly measured/unmeasured - 90.9% of the inventory,
_ difficult to measure - 6.8% of the inventory, and
_ non-amenable to measurement - 2.3% of the inventory.

A logical consequence of the MAP was the detailed breakdown of the HEU inventory at LLNL. The most predominate form descriptions in the MAP HEU inventory and the approximate percentage by mass of the total each represents are summarized in Table 1.

Table 1. Most predominate from descriptions in the LLNL HEU inventory.

Item form description	Approximate percentage of inventory by mass
Metal criticality assembly shells	40%
Metal (other forms)	18%
Oxides	5%
Mixed U/Pu oxides	18%
Assemblies	17%
Fuel elements and plates	1%
Miscellaneous parts	1%

Initial Instrument Calibration Attempts

To obtain the best possible measurements on the poorly measured/unmeasured material, HEU metal and oxide certified reference material (CRM) standards were needed for calibration of LLNL's neutron-based non-destructive assay (NDA) instruments: active-well coincidence counter (AWCC), neutron multiplicity counter (NMC) with active mode insert and passive-active neutron (PAN) shuffler. In anticipation of a lengthy development time for both metal and oxide CRM standards, LLNL began to examine the possibility of developing working standards from the HEU inventory items that were currently on site. Because of the geometry dependence of neutron instruments in general, the variety of geometries represented in the LLNL HEU inventory, the mass fraction of the HEU inventory contained in hemispherical criticality shells, the existence of a Los Alamos National Laboratory (LANL) HEU metal hemispherical criticality shell calibration curve for the AWCC, and the existence of a LANL HEU oxide calibration curve for the PAN shuffler, the initial focus on working standards at LLNL was limited to hemispherical criticality shells.

During the initial calibration attempts, a large fraction of the HEU metal (_72% by mass) and oxide (_85% by mass) inventory was measured in the PAN shuffler and the data archived. The overwhelming majority of these items were measured as single entities and not as part of a multi-pack. A cursory examination of the data results exhibited a general correlation with mass, enrichment, size, and shape.

Current Status of Standards

A DOE-HQ sponsored survey of the DOE complex by New Brunswick Laboratory (NBL) has resulted in their preparation of CRM 149, a set of 93% enriched U_3O_8 standards that are projected for delivery to LLNL during September of 1999. However, CRM standards for HEU metal are neither in preparation at NBL, nor are they expected to be prepared within the foreseeable future. Consequently, HEU metal working standards must be developed if that portion of the LLNL poorly measured/unmeasured inventory is ever to be reclassified.

Instrument Calibration and Measurement Plan

Overall Approach

LLNL will develop a set of HEU metal and oxide calibration curves for the PAN shuffler that are functions of mass, enrichment, size, and shape. In the case of the HEU metal, working standards will be created from selected item shapes within that inventory. Selection of the HEU metal item shapes will follow a graded approach and be based on the most attractive with respect to mass, size, and enrichment. For item shapes, sizes, and enrichments not represented in the selection process, computational modeling will be required to establish the differences in instrument response and provide corrections to the basic calibration data. In the case of the HEU oxide, the CRM 149 standards currently in preparation at NBL will provide the basic calibration data. For container shapes and sizes and item enrichments not represented by the CRM 149 standards, computational modeling will be required to relate the differences in instrument response and provide corrections to the basic calibration data. In addition, because of the difference in chemical form between the CRM 149 standards (U_3O_8) and the LLNL HEU oxide items (UO_2), computational modeling will also be required to relate the differences in instrument response and provide corrections to the basic calibration data.

Once the appropriate HEU metal and oxide calibration curves are established, the order of precedence for the measurement plan will be as follows:

_ reprocess all archived data for HEU metal and oxide items previously measured as single entities or in multipacks where the items were of common shape and common enrichment,

_ measure all remaining single entity HEU metal and oxide items or multipacks where the items are of common shape and common enrichment, and

_ repackage all remaining HEU metal and oxide multipacks into single entity items or multipacks of common shape and common enrichment and measure accordingly.

Detailed Approach

More detailed discussions of the instrument calibration and measurement plan are provided in the following sections along with an estimate of the time required to complete each phase.

Inventory Survey

The LLNL poorly measured/unmeasured HEU metal and oxide inventory will be updated to include any material transfers or receipts since the MAP HEU inventory was initially reported. As part of the update process, the HEU metal and oxide inventories will be segregated into the following categories: single items previously measured in the PAN shuffler, single items not previously measured in the PAN shuffler, multipack items previously measured in the shuffler, and multipack items not previously measured in the shuffler. In addition, the measured and unmeasured multipack categories will be further segregated into the following categories: common shape and common enrichment, common shape and mixed enrichment, mixed shape and common enrichment, and mixed shape and mixed enrichment. Within all categories, the items will be rank sorted in the order of element mass, shape description, and enrichment.

HEU Metal Working Standard Selection

Once the inventory survey is complete, HEU metal items can be selected for use as working standards. A graded approach will be employed in the selection process. Those metal items with the most element mass and the most common shape description will receive the highest ranking. Next in order will be metal items with the second most element mass and the most common shape description. This ordering process will continue until all metal items have been ranked. Items selected for working standards will come from the three most common shapes with the highest ^{235}U mass. To evaluate the effect of ^{235}U enrichment on instrument response, a maximum of three enrichments will be selected from the range of enrichments within each shape description group. Whether these selected enrichments will reflect the maximum, minimum, and mid-range; the three most common; or some other combination cannot be specified at this time but must await completion of the inventory survey. Overall, LLNL expects to identify a total of no more than nine HEU metal items for use as working standards.

Measurement of HEU Metal Working Standard Mass and Enrichment

Once the HEU metal working standards have been identified, the mass and ^{235}U isotopic enrichment of each will be established by measurement. Each working standard will be moved to a uranium glove box where it will be unpackaged, weighed by electronic balance, measured by enrichment meter, and repackaged. The mass of the working standard and the uncertainty in that mass will be determined from the average of five or more separate weighings and three times the associated standard deviation in that average (3 _). The ^{235}U isotopic enrichment of the working standard and the uncertainty in that enrichment will be determined from the average of eight or more separate enrichment meter measurements taken over the surfaces of the working standard and three times the associated standard deviation in that average (3 _). To avoid contamination of the enrichment meter and associated electronics, measurement of each working standard will be accomplished through one of the glove box gloves. As such, the enrichment meter will be calibrated with a representative piece of glove material to account for the associated shielding effects.

Measurement of HEU Metal Working Standards

The number of previously measured and archived results for each HEU metal working standard will be determined and the results reviewed. While the software includes a decay correction factor that increases the delayed neutron count rate, with nearly three years having elapsed since the previous measurements were made, at least one new measurement will be performed on each working standard to examine the effect of ^{252}Cf decay (t_ = 2.646 years) on the measurement precision. If there are at least five acceptable archived measurements for each working standard, only the one additional measurement will be required for each to complete the development of the calibration curves. However, if there are not at least five acceptable archived measurements for each working standard, additional measurements will be required. Both archived and newly acquired data will then be processed with the appropriate working standard mass and ^{235}U isotopic enrichment data.

Preparation of HEU Metal Working Standard Calibration Curves and Measurement Controls

Calibration curves will be generated from the HEU metal working standard data for the three most common shape descriptions over the ^{235}U enrichment range of interest. It is anticipated that these calibration curves will be expressed in a like or similar manner to those generated from earlier studies with the PAN shuffler at LLNL and LANL. Uncertainties associated with the mass predicted by the calibration curves will be derived from a statistical analysis of the fitted data.

Once the calibration curves are generated and their associated uncertainties are derived, one or more working standards will be selected from each of the three most common shape descriptions for measurement control. Alarm and warning limits will be established at two and three times the standard deviation (2 and 3 _) in the average of the appropriate measured working standard mass, respectively.

Measurement of HEU CRM 149 Oxide Standards

The CRM 149 oxide standard set being prepared by NBL is expected to consist of a blank unit plus a maximum of six units that contain 0.5 kg, 1.0 kg, 1.5 kg, 2.0 kg, 3.0 kg, and 4.0 kg of 93% enriched U_3O_8. A minimum of five replicate measurements will be required for each CRM 149 standard.

Preparation of HEU Oxide CRM Standard Calibration Curves and Measurement Controls

A calibration curve will be generated from the CRM 149 oxide standard data over the ^{235}U mass range of the standards. It is anticipated that this calibration curve will be expressed in a like or similar manner to those generated from earlier studies with the PAN shuffler at LLNL and LANL. Uncertainties associated with the mass predicted by the calibration curves will be derived from a statistical analysis of the fitted data.

Once the calibration curve is generated and its associated uncertainties are derived, one or more CRM 149 standards will be selected for measurement control. Alarm and warning limits will be established at two and three times the standard deviation (2 and 3 _) in the average of the appropriate measured CRM 149 standard mass, respectively.

Computational Modeling of Unrepresented HEU Metal and Oxide Items

Only the three most common HEU metal shape descriptions and three associated ^{235}U enrichments will be represented by the HEU metal working standards. Whereas none of HEU oxide shape descriptions (i.e., containers) are expected to be represented by the CRM 149 oxide standards. For those HEU metal and oxide shape descriptions (item or container), sizes, and item ^{235}U enrichments not represented by the working or CRM 149 standards, computational modeling will be performed to establish the differences in instrument response and provide corrections to the basic calibration data. In addition, computational modeling will be performed to establish the difference in instrument response due to the difference in chemical form between the CRM 149 standards (U_3O_8) and the LLNL HEU oxide items (UO_2) and provide corrections to the basic calibration data.

The PAN shuffler instrument will be modeled with a point-wise cross-section Monte Carlo code such as MCNP. Models of the HEU metal working standards, CRM 149 oxide standards, and unrepresented shape descriptions as single entity items will be created and the detector response calculated for each. For a given single entity item shape description (item or container), sensitivity studies will be performed to determine the variability in detector response with respect to (1) size, (2) item ^{235}U enrichment, (3) packaging, (4) in the case of the HEU metals, possible geometric orientation (e.g., top of can—horizontal and vertical, middle of can—horizontal and vertical, bottom of can—horizontal and vertical), and (5) in the case of HEU oxides, chemical form (i.e., U_3O_8 versus UO_2) and moisture content. For multipacks containing items of a common shape description, additional sensitivity studies will be performed to determine the variability in detector response with respect to the orientation of each item and its container. Comparisons will be made between the unrepresented shape descriptions and associated sensitivity results and the calculated detector responses to the HEU metal working standards and oxide CRM standards. From these comparisons, normalization factors will be derived for the unrepresented shape descriptions and ^{235}U enrichments in the inventory.

Since the results derived from MCNP are ultimately to be used to establish calibration curves, the smaller their standard deviation the better. However, with the MCNP results being statistical, the level of accuracy required in the final result will dictate the length of time required to analyze each item (e.g., for a given level of accuracy, an item with very low mass could require a week of computation time while an item of very high mass could require no more than a few hours).

Development of Correction Factors for the HEU Metal and Oxide Calibration Curves

Comparisons will be made between the measured and calculated results for the HEU metal working standards and CRM 149 oxide standards and mass normalization factors derived for each. These mass normalization factors will be applied to the calibration curves to produce an effective family of calibration curves for all shape descriptions analyzed as a function of ^{235}U enrichment. Like the calibration curves, uncertainties associated with the mass predicted by these effective families of calibration curves will be derived from a statistical analysis of the fitted data.

Reprocess Selected HEU Metal and Oxide Archive Data

All archived data for those HEU metal and oxide items that were previously measured as single entities or in multipacks where the items were of common shape and enrichment will be reprocessed.

Measure Selected HEU Metal and Oxide Items

All remaining single entity HEU metal and oxide items or multipacks where the items are of common shape and enrichment will be measured.

Repackage and Measure Selected HEU Metal and Oxide Items

All remaining HEU metal and oxide multipacks will be repackaged in a uranium glove box into single entity items or multipacks of common shape and common enrichment and measured accordingly.

Total Time Estimate

Table 2. summarizes the best case and worst case estimates of the time required to complete each phase. Under the best case scenario of dedicated individuals and high priorities assigned to each phase, approximately four years and two months will be required to complete the effort. However, in the event that neither dedicated individuals nor high priorities are assigned, approximately nine years and two months could be required to complete the effort.

Table 2. Best case and worst case estimates of the time required to complete each phase.

Project phase	Time required to complete (months)	
	Best case	**Worst case**
1. Inventory survey	3	3
2. HEU metal working standard selection	1	1
3. Measurement of HEU Metal working standard mass and enrichment	0.25	0.75
4. Measurement of HEU metal working standards	0.75	3
5. Preparation of HEU metal working standard calibration curves and measurement controls	1	1
6. Measurement of HEU CRM 149 oxide standards	0.50	2
7. Preparation of HEU oxide CRM standard calibration curves and measurement controls	1	1
8. Computational modeling of unrepresented HEU metal and oxide items	36	72
9. Development of correction factors for the HEU metal and oxide calibration curves	2	2
10. Reprocess selected HEU metal and oxide archived data	1.50	11
11. Measure selected HEU metal and oxide items	0.75	4
12. Repackage and measure selected HEU and metal oxide items	2	10
Total	50	111